Dephasing of two-spin states by electron-phonon interaction in semiconductor nanostructures: Spin-boson model with a dissipative reservoir

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We study electron-phonon interaction induced decoherence between two-electron singlet and triplet states in a semiconductor double quantum dot using a spin-boson model. We investigate the onset and time evolution of this dephasing, and study its dependence on quantum dot parameters such as dot size and double dot separations, as well as the host materials (GaAs and Si). We find that electron-phonon interaction causes an initial Gaussian decay of the off-diagonal density matrix element in the singlet-triplet Hilbert space, and a long-time exponential decay originating from phonon relaxation.

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In the past few years, significant experimental progresses in the study of semiconductor spin qubits [1, 2, 3, 4, 5] have reconfirmed electron spins as one of the leading candidates for the building block of a solid state quantum information processor. Recent theoretical studies have also clarified single spin decoherence channels and their relative importance in semiconductor quantum dots [6, 7, 8, 9, 10, 11], with hyperfine interaction with environmental nuclear spins identified as the main culprit for electron spin decoherence. As experimental studies shift toward controlled coupling of spin qubits, quantum coherence properties of multiple-spin states are naturally the next important theoretical problem, whose solution could go a long way in determining whether spin qubits in semiconductor nanostructures indeed have desired scalability for a practical quantum computer.

Decoherence of two-spin states in a coupled double quantum dot is crucial to the operation of an exchange-based spin quantum computer architecture [12]. Since nuclear spins are the main source of single spin decoherence in quantum dots, existing studies have focused on the decohering effects of the nuclear spins [13, 14]. In addition, since exchange coupling is electrostatic in nature, exchange-coupled electrons are vulnerable to charge noise and other orbital fluctuations that have an electrical signature. For example, we have shown [15] that charge fluctuations lead to pure dephasing by introducing noise into interdot barrier and/or interdot voltage bias.

Here we study decoherence effects of electron-phonon interaction on two-spin states in semiconductor nanostructures, based on the consideration that lattice vibrations produce local electrical polarizations and could therefore affect the two-spin singlet and triplet states (two-spin eigenstates in the absence of spin-orbit interaction) for exchange-coupled electrons. We focus on symmetrically coupled (i.e. no inter-dot voltage bias) quantum dots in GaAs and Si, and P donors in Si, all regarded as promising candidates for qubits in spin-based quantum information processing. Electron-phonon interaction is not spin-dependent and does not lead to direct transitions between singlet and triplet states. Consequently there is no phonon-induced relaxation between the singlet and triplet states in the absence of spin-orbit interaction. Our focus is on pure dephasing effect of electron-phonon interaction in a two-electron double dot. Specifically, we obtain the effective interaction Hamiltonian, identify the most important types of electron-phonon interaction, clarify the dynamics of dephasing, and quantify its time scale in GaAs and Si.

The general electron-phonon interaction Hamiltonian in a semiconductor takes the form [16]

$$H_{ep} = \sum_{\mathbf{q},\lambda} M_\lambda(\mathbf{q})\rho(\mathbf{q})(a_{\mathbf{q},\lambda} + a^\dagger_{-\mathbf{q},\lambda}) \,, \qquad (1)$$

where $a_{\mathbf{q},\lambda}$ and $a^\dagger_{-\mathbf{q},\lambda}$ are phonon annihilation and creation operators with lattice momentum $\mathbf{q}$ and branch index λ, and $\rho(\mathbf{q})$ is the electron density operator. For an electron to act as a spin qubit, its orbital degree of freedom needs to be frozen in the ground state. When two spin qubits are exchange-coupled in a symmetric double quantum dot, their orbital states are symmetric or antisymmetric if their spin state is singlet ($|\uparrow\downarrow - \downarrow\uparrow\rangle/\sqrt{2}$) or triplet ($|\uparrow\downarrow - \downarrow\uparrow\rangle/\sqrt{2}$, $|\uparrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$). Within the Heitler-London approximation, the two spatial wave functions can be written as

$$\begin{aligned} \psi_S &= \frac{1}{\sqrt{2(1+S^2)}}|L(1)R(2) + R(1)L(2)\rangle \,, \\ \psi_{AS} &= \frac{1}{\sqrt{2(1-S^2)}}|L(1)R(2) - R(1)L(2)\rangle \,, \end{aligned} \qquad (2)$$

where L and R refer to the ground single-electron orbital states in the two dots, $S = \langle L|R\rangle$ is the overlap integral, and 1 and 2 are indices for the two electrons. We can now project the electron-phonon interaction into the singlet-triplet Hilbert space. All three triplet states have the

same orbital wave function and cannot be differentiated by electron-phonon interaction. The Hilbert space of interest is thus only two-dimensional, with the corresponding basis states $\frac{1}{\sqrt{2(1+S^2)}}|L(1)R(2)+R(1)L(2)\rangle \times \frac{1}{\sqrt{2}}|\uparrow\downarrow - \downarrow\uparrow\rangle$ and $\frac{1}{\sqrt{2(1-S^2)}}|L(1)R(2)-R(1)L(2)\rangle \times \frac{1}{\sqrt{2}}|\uparrow\downarrow + \downarrow\uparrow\rangle$. Since the Hamiltonian has no spin-dependence, the 2×2 electron-phonon interaction Hamiltonian is diagonal:

$$H_{eff} = \sum_{\mathbf{q},\lambda} M_\lambda(\mathbf{q}) A_\phi \sigma_z (a_{\mathbf{q},\lambda} + a^\dagger_{-\mathbf{q},\lambda}) \,, \qquad (3)$$

where σ_z is a Pauli matrix in the two-dimensional two-electron Hilbert space (it is not for single electron spins), and the charge distribution difference A_ϕ is given by

$$A_\phi = \frac{1}{2}[\langle\psi_{AS}|\rho(\mathbf{q})|\psi_{AS}\rangle - \langle\psi_S|\rho(\mathbf{q})|\psi_S\rangle] \,, \qquad (4)$$

where $\rho(\mathbf{q}) = e^{i\mathbf{q}\cdot\mathbf{r}_1} + e^{i\mathbf{q}\cdot\mathbf{r}_2}$ [17]. A_ϕ is completely determined by the charge distribution difference between the two-electron singlet and triplet states. Specifically, in a singlet state (symmetric wave function) the electrons tend to distribute themselves across the two dots to minimize their kinetic energy, while in a triplet (antisymmetric wave function) the two electrons avoid each other to minimize the Coulomb interaction. For the symmetric double dot we study here, the singlet state has larger charge density in between the two dots, while the triplet has larger charge density at the far ends of the double dot. The resulting difference in charge distribution has a finite electrical quadrupole moment and gives A_ϕ its $\mathbf{q}$-dependence.

The effective electron-phonon interaction Hamiltonian of Eq. (3) is a typical spin-boson Hamiltonian that leads to decay in the off-diagonal element of the 2×2 density matrix [18]:

$$\rho_{ST}(t) = \rho_{ST}(0) e^{-B^2(t)} \,, \qquad (5)$$

where the dephasing factor is positive definite:

$$B^2(t) = \frac{2V}{\pi^3\hbar^2}\int d^3\mathbf{q} \frac{|M(\mathbf{q})A_\phi(\mathbf{q})|^2}{\omega_\mathbf{q}^2} \sin^2\frac{\omega_\mathbf{q} t}{2} \coth\frac{\hbar\omega_\mathbf{q}}{k_B T} \,. \qquad (6)$$

Here $\omega_\mathbf{q}$ is the angular frequency of the phonons in mode $\mathbf{q}$. The derivation of this dephasing formula account for the fact that the bosonic reservoir is in a thermal equilibrium before getting into contact with the spin [18]. However, it does not account for the fact that the bosonic modes may be dissipative. Indeed, an ordinary spin-boson calculation of dephasing generally assumes a spectral density of the form $1/f^\alpha$, with the implicit assumption that bosonic modes with a vanishing spectral density at low frequency does not contribute to dephasing in any significant way. In the present study, acoustic phonons do have a vanishing density of state at low frequency. On the other hand, we also know that these phonons also decay very fast, with a time scale as short as 10-100 ps (by anharmonicity induced phonon decay and by irreversibly propagating out of the nanostructure). By adding such a phonon decay channel (for simplicity, we assume all phonon modes have the same decay rate γ), we obtain a modified expression for the dephasing of the off-diagonal density matrix element:

$$\begin{aligned}
\rho_{ST}(t) &= \rho_{ST}(0) e^{-B_1^2(t) - B_2^2(t)} \,, \\
B_1^2(t) &= \frac{V}{\pi^3\hbar^2}\int d^3\mathbf{q} \frac{|M(\mathbf{q})A_\phi(\mathbf{q})|^2}{\omega_\mathbf{q}^2 + (\gamma/2)^2} \\
&\quad \times \left\{ \frac{\omega_\mathbf{q}^2 - (\gamma/2)^2}{\omega_\mathbf{q}^2 + (\gamma/2)^2}\left(1 - e^{-\frac{\gamma}{2}t}\cos\omega_\mathbf{q} t\right) \right. \\
&\quad \left. - \frac{\omega_\mathbf{q}\gamma/2}{\omega_\mathbf{q}^2 + (\gamma/2)^2} e^{-\frac{\gamma}{2}t}\sin\omega_\mathbf{q} t \right\} \coth\frac{\hbar\omega_\mathbf{q}}{k_B T} \,, \qquad (7) \\
B_2^2(t) &= \frac{V}{\pi^3\hbar^2}\int d^3\mathbf{q} \frac{|M(\mathbf{q})A_\phi(\mathbf{q})|^2}{\omega_\mathbf{q}^2 + (\gamma/2)^2} \left(\frac{\gamma}{2} t\right) \coth\frac{\hbar\omega_\mathbf{q}}{k_B T} \\
&= \Gamma_{ST} t \,. \qquad (8)
\end{aligned}$$

At the limit that phonon decay rate $\gamma \to 0$, $B_1^2(t) \to B^2(t)$ while $B_2^2(t) \to 0$. For a finite γ, corresponding to a dissipative phonon reservoir, we obtain an additional exponential decay of the off-diagonal density matrix element in Eq. (8) compared to the non-dissipative reservoir result of Eq. (6). The rate of this exponential decay Γ_{ST} is proportional to both the phonon decay rate γ and the geometric factors that determine the dephasing factor $B_1^2(t)$.

Let us now examine the dynamical behaviors of the dephasing factors $B^2(t)$ and $B_2^2(t)$. In Fig. 1 we show the typical behavior of the dephasing factor $B^2(t)$ in the absence of phonon decay for various types of electron-phonon interactions in GaAs and Si. There are two interesting features all the curves in Fig. 1 share. At very short times ($t \ll 1$ ps), the increase of $B^2(t)$ is quadratic, which originates from Taylor expansion of the $\sin^2\omega_\mathbf{q} t/2$ factor in the integrand at the small t limit. At long times all the curves saturate, which means that dephasing does not increase with time anymore, so that it corresponds more to a finite loss of contrast than the conventional complete decay of off-diagonal density matrix elements. The transition between the quadratic increase and the saturation happens between 1 and 10 ps for double dots with dot separation of about 40 nm and somewhat shorter for P pairs because this time is essentially determined by the interdot distance divided by the speed of sound ($\sim 8\times 10^3$ m/s in Si and 3.7×10^3 m/s in GaAs). Mathematically the long-time saturation can be understood by writing $2\sin^2\omega_\mathbf{q} t/2$ as $1-\cos\omega_\mathbf{q} t$. Since acoustic phonon spectrum is continuous, the cosine term leads to a vanishing contribution to the integral at large times, which leaves the dephasing factor determined by a constant integral that is independent of time. Physically,

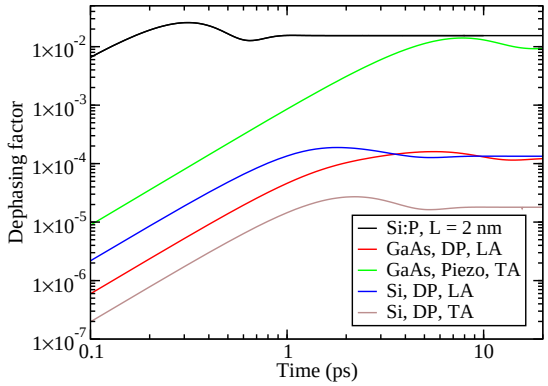

FIG. 1: (Color online) Two-spin dephasing in a double quantum dot induced by a non-dissipative phonon reservoir. The solid curve represents $B^2(t)$ for a P pair in Si separated by 4 nm (The Bohr radius for Si:P is $\lesssim$ 2 nm). All the other curves are for double quantum dots with an interdot separation of 40 nm and single dot orbital radius of 20 nm. More specifically, the red dotted curves are for deformation potential (DP) coupling to longitudinal acoustic (LA) phonons in GaAs; the green dashed curve is for piezoelectric (PE) coupling to TA phonons in GaAs; the blue dot-dashed curve is for DP coupling to LA phonons in Si; and the brown dot-dot-dashed curve is for DP coupling to TA phonons in Si.

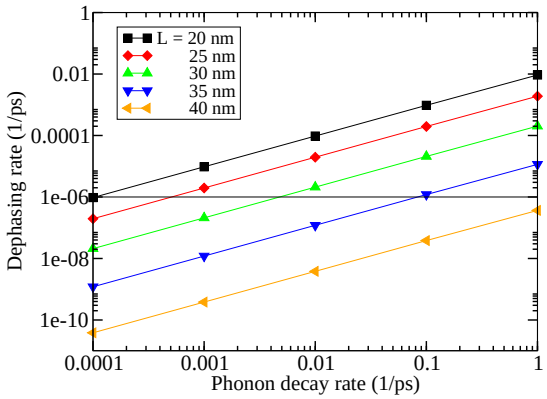

FIG. 2: (Color online) Phonon induced two-spin dephasing rate as a function of phonon decay rate in a GaAs double dot for various interdot separation distance. The horizontal line is drawn at a dephasing time of 1 μs, approximately the decoherence times measured in Refs. [1, 2]. The single dot wave function radius for all the data is 20 nm.

this saturation is due to the fact that long-time dephasing is determined by the low-frequency part of the spectrum of the bosonic reservoir, while phonon density of state vanishes quadratically at low frequency. In other words, non-dissipative acoustic phonons simply form an inefficient dephasing reservoir as compared to other charge fluctuation reservoirs such as fluctuating charge traps, which have a $1/f$ spectral density.

The magnitudes of the saturated dephasing shown in Fig. 1 give a clear sense of the relative importance of various types of electron-phonon interactions. Specifically, in GaAs the piezoelectric (PE) coupling to transverse acoustic (TA) phonons produces the strongest dephasing effect, while in Si the deformation potential (DP) coupling to longitudinal acoustic (LA) phonons is the most important. Here the phosphorus dimer has the strongest dephasing because it has one-order-of-magnitude smaller inter-donor distance, so that the dominant contribution to its dephasing comes from higher energy phonons, which have higher density of states, compared to those for quantum dots.

When phonon decay is included, the most important additional effect is the added exponential dephasing $e^{-\gamma_{ST}t}$. In Fig. 2 we plot the dephasing rate Γ_{ST} as a function of the interdot distance and the phonon decay rate γ. As an example we plot the dephasing rate in a GaAs double dot due to PE coupling to TA phonons. This is by far the strongest dephasing channel in either GaAs or Si, as clearly indicated in Fig. 1. Our results show that for faster phonon decay, we need to keep the interdot distance sufficiently large in order to obtain longer singlet-triplet dephasing time.

In Fig. 3 we plot the phonon-induced dephasing rate between singlet and triplet states in double dots in both GaAs (diamond symbols) and Si (triangular symbols) as functions of the interdot distance L. The strong dependence on L originates from the fact that charge distribution difference between the two-electron singlet and triplet states is directly dependent on interdot wave function overlap: The smaller the overlap, the smaller the difference in charge distribution, and the smaller the phonon-induced dephasing. The Si data is about two orders of magnitude smaller than in GaAs, consistent with what is shown in Fig. 1, and is determined by the fact that deformation potential interaction in Si is simply a weaker interaction than piezoelectric interaction in GaAs.

The magnitude of phonon-induced dephasing to two-spin states in a double dot depends directly on how fast the phonons themselves relax. If phonon relaxation is dominated by internal mechanisms such as phonon anharmonicity [19], with a time scale much longer than nanoseconds, the phonon-induced dephasing would be relatively slow, with a time scale above μs. On the other hand, if phonons escape the nanostructure rapidly, in the order of 10 ps to 1 ns, the double dot will need to be well separated for phonon-induced dephasing to be sufficiently slow. Whether this dephasing is slow enough, we need to compare it with the speed of gating.

In Fig. 4 we plot the two-spin merit figure $\mathcal{M}$ as a function of the interdot distance for double dots in GaAs. Here the merit figure is defined as the ratio between a typical exchange gate time given by $\hbar/J$ (J is the exchange splitting) and the two-spin decay time given by $1/\gamma_{ST}$: $\mathcal{M} = \hbar\gamma_{ST}/\mathcal{J}$. The exchange splitting J is calculated

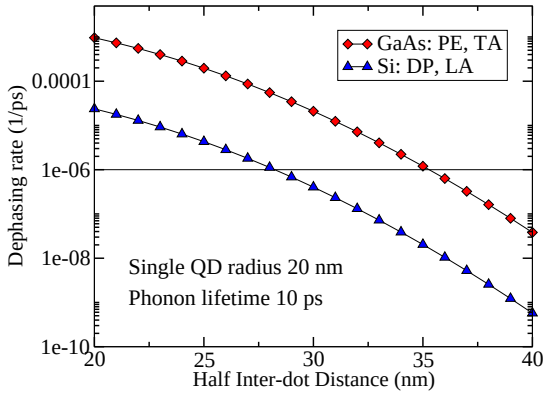

FIG. 3: (Color online) Phonon-induced two-spin dephasing rate as a function of the interdot separation for a GaAs and a Si double quantum dot. The horizontal line is again drawn at a dephasing time of 1 μs. For GaAs the curve is for piezoelectric (PE) coupling to TA phonons, while for Si it is for deformation potential (DP) coupling to LA phonons.

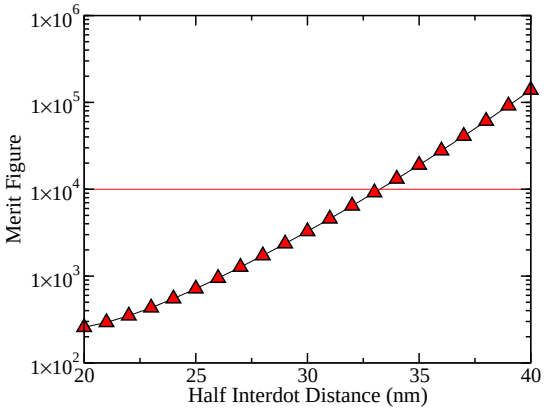

FIG. 4: (Color online) Merit figure based on phonon-induced dephasing of two-spin states in a GaAs double dot as a function of half interdot distance. We draw a line at 10^4 as the nominal threshold for fault tolerant quantum computation. Therefore the double dot (with single-dot wave function radius at 20 nm) should be kept apart further than 70 nm.

within the Heitler-London model with a quartic confinement potential [20]. The radius of the single dot electron wave function is 20 nm. We choose a phonon decay time of 10 ps as a worst case scenario. The increase of the merit figure at larger inter-dot distance reflects the fact that the exchange splitting and the phonon-induced dephasing have different dependence on the interdot overlap integral S: $J \sim S^2$, while $\gamma_{ST} \sim S^4$. The results shown in this figure reveal that for a two-dot exchange gate to operate with a low error rate, slower operation with smaller interdot overlap is preferable. We do not have any data for Si quantum dot in this figure. Calculating exchange interaction in a Si double dot requires much more sophisticated quantum chemical approaches than a simple Heitler-London approximation [21, 22] because in Si the interaction effect is stronger compared to GaAs (larger effective mass and smaller dielectric constant), so that Heitler-London approximation does not adequately account for the two-electron correlation. For the current evaluation, it is sufficient to point out that Fig. 3 above indicates that phonon-induced dephasing is about two orders of magnitude weaker in Si than in GaAs, while exchange coupling should only be somewhat smaller than in GaAs. Therefore overall there should a gain of at least one order of magnitude in the merit figure when moving from GaAs to Si.

We have shown here that while the phonon reservoir is treated as a continuum of states, in the absence of phonon relaxation it does not lead to complete dephasing of the two-spin states. This somewhat counter-intuitive result can be understood from two different perspectives. One is the quadratically vanishing phonon density of state at low frequency, as we mentioned previously. The other is the fact that the electron-phonon interaction matrix element vanishes at high phonon frequency for the quantum dot orbital states. For example, with a Gaussian envelope function, the matrix element $\langle L|e^{ikx}|L\rangle \sim e^{k^2a^2/4}$ where k is the phonon wave vector and a is the wave function width. Thus for large k these matrix elements vanish rapidly. Therefore the part of the phonon spectrum that actually contributes to spin dephasing is relatively small. For a quantum dot with ground state wave function radius in the order of 20 nm, only those acoustic phonons with energy below 1 meV are relevant. In a realistic lattice, the electron states are always dressed by the phonons. What the results in Fig. 1 show is that the dressed states still have their majority spectral weight in the bare two-electron states. On the other hand, when phonons can relax completely, they bring the electrons into contact with an even larger reservoir that is not closed, so that the two-spin states can dephase completely.

In our calculations we consider double quantum dots that are symmetric: the two dots are the same in size and are not voltage biased, in the traditional Loss-DiVincenzo configuration for implementing an exchange gate. In some recently studied double dot systems a voltage bias is applied between the two dots so that the system is close to the bias point where two-dot singlet state and one of the double-occupied singlet states are degenerate [1]. In this configuration the charge distribution difference between the ground singlet and triplet states are more dramatic because the singlet state has a finite spatial component of doubly occupied state while the triplet does not have such a component. In our calculation this should lead to a much larger A_ϕ defined in Eq. (4), which in turn leads to a larger spin-boson coupling matrix element. Accordingly the phonon-induced dephasing should in general be faster than in the symmetric situation. A complete calculation for this situation is beyond

the scope of the current paper.

In conclusion, we have studied phonon-induced dephasing between two-electron singlet and triplet spin states in a semiconductor double quantum dot. We find that this dephasing is important for tightly coupled double dots, especially when phonon decay is taken into consideration. We re-derive the expression for dephasing in the spin-boson model with a dissipative reservoir, and quantify the the two-spin dephasing in both GaAs and Si double dots.

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